Scaling constant and its determination from simultaneous measurements of light reflection and methane adsorption by snow samples

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Received June 22, 2006; revised August 27, 2006; accepted August 29, 2006;
posted September 6, 2006 (Doc. ID 72265); published October 26, 2006

We describe a fast and accurate method for the determination of the specific surface area of snow samples based on the measurements of the snow reflection function at a single wavelength and geometry. The method is less sensitive to the assumed shape of particles as compared with other techniques. The concept of the snow scaling constant is introduced, and its value is derived from simultaneous measurements of light reflectance and methane adsorption. © 2006 Optical Society of America

OCIS codes: 280.0280, 120.0280.

Photometric techniques used to measure the effective snow grain size $d=6V/\Sigma$, where V is the average volume of crystals and Σ is their average surface area, are based on the assumption that grains have spherical shapes.[1–5] This enables the determination of the specific surface area (SSA) of snow defined as $\sigma=\Sigma/\rho V$, where $\rho=0.917\ \mathrm{g/cm^3}$ is the density of ice, which is of importance for snow chemistry studies.[6,7] Note that it follows $d=6/\rho\sigma$. Usually the snow reflection function R at one or several wavelengths is measured, and the value of d is inferred by fitting radiative transfer calculations to measurements for semi-infinite snow layers in the near IR.[5] The radiative transfer models used are very remote from reality, representing snow as a collection of ideal ice spheres placed at large distances from one another. It is not very difficult to introduce particles of other shapes in the theoretical model.[8–10] However, then the problem still remains because grains have very complicated geometry and cannot be described by simple, single, regular forms or by their combinations.[7]

In this Letter a simple and accurate method to determine the snow grain size and, therefore, the specific surface area of snow is described. The method is based on the reflection function measurements at a single wavelength and for a fixed observation geometry. The concept of the scaling constant f is introduced. The technique to measure f is proposed and applied to simultaneous measurements of the pair (σ,R) for natural snow in the field.[7]

Consider a plane-parallel semi-infinite slab filled by irregularly shaped weakly absorbing ice particles. The reflection function of such a slab is described by the following analytical equation[9]:

$$R(\vartheta_0,\vartheta,\varphi)=R_0(\vartheta_0,\vartheta,\varphi)\exp[-4sU(\vartheta_0,\vartheta,\varphi)], \quad (1)$$

where $\vartheta_0,\vartheta,\varphi$ are incidence, observation, and azimuth angles, respectively, $R_0(\vartheta_0,\vartheta,\varphi)$ is the reflection function of the same layer but at no absorption (single-scattering albedo $\omega_0=1$),

$$s=\sqrt{\frac{(1-\omega_0)}{3(1-\omega_0 g)}} \quad (2)$$

is the similarity parameter, g is the asymmetry parameter,

$$U(\vartheta_0,\vartheta,\varphi)=\frac{u_0(\vartheta_0)u_0(\vartheta)}{R_0(\vartheta_0,\vartheta,\varphi)} \quad (3)$$

is the angular function, and $u_0(\vartheta_0)$ is the escape function,[9] which can be approximated with an accuracy of better than 2% as

$$u_0(\vartheta_0)=\frac{3}{7}(1+2\cos\vartheta_0) \quad (4)$$

for particles of arbitrary sizes and shapes[9] at angles smaller than 75°. The weak sensitivity of the escape function to the shape and size of particles is due to the fact that $u_0(\vartheta_0)$ describes the diffusion of photons from the infinite depth inside the nonabsorbing scattering medium to the upper surface of the medium.[9] Then the peculiarities related to single-scattering laws of nonspherical particles having different shapes are largely washed out. This is approximately true for the reflection function of a nonabsorbing medium $R_0(\vartheta_0,\vartheta,\varphi)$ as well. In particular, reflection functions of semi-infinite nonabsorbing turbid media with hexagonal and fractal particles are close to each other.[11] Therefore the shape of particles enters the theory mostly through the similarity parameter s, which can be easily determined from Eq. (1):

$$s=\frac{1}{4U(\vartheta_0,\vartheta,\varphi)}\ln\left\{\frac{R_0(\vartheta_0,\vartheta,\varphi)}{R(\vartheta_0,\vartheta,\varphi)}\right\}, \quad (5)$$

where R is the measured reflection function and R_0 is the calculated one (e.g., in the model of fractal nonspherical grains[8,9]). Therefore the determination of the similarity parameter s is much less affected by the assumption on the shape of particles as compared with that of d. The relationship s with d ($d\sim 1-\omega_0$ for

0146-9592/06/223282-3/$15.00

weakly absorbing large grains as $\omega_0 \to 1$, see below) is, however, heavily affected by the assumption on the shape of particles. In particular, g in Eq. (2) is around 0.9 for nonabsorbing ice spheres, and it is just 0.75 for nonabsorbing fractal particles independent of their size,[9] if $d \gg \lambda$, where λ is the wavelength.

To solve the problem we propose the following technique. We use the fact that the absorption cross section C_{abs} of arbitrarily shaped weakly absorbing large grains is proportional[9] to their volume v: $C_{\text{abs}} = B\gamma v$, where $\gamma = 4\pi\chi/\lambda$, χ is the imaginary part of the refractive index of ice, and B is *a priori* unknown shape-dependent constant having the property $B \to 1$ as the refractive index $n \to 1$. The extinction cross section is proportional to the geometrical cross section of the particle S with the coefficient of proportionality equal to 2 independent of the shape[9]: $C_{\text{ext}} = 2S$. The single-scattering albedo is defined as $\omega_0 = 1 - K_{\text{abs}}/K_{\text{ext}}$, where $K_{\text{abs}} = N\langle C_{\text{abs}}\rangle$, $K_{\text{ext}} = N\langle C_{\text{ext}}\rangle$, N is the number of particles in a unit volume, and $\langle\ \rangle$ means averaging with respect to the size and shape of particles. Therefore it follows as $\omega_0 \to 1$

$$\omega_0 = 1 - 2B\gamma\frac{V}{\Sigma}, \tag{6}$$

where $V = \langle v\rangle$, and we used the fact that $\Sigma = 4\langle S\rangle$ for randomly oriented convex particles.[9] Therefore one obtains from Eq. (2), for particles of arbitrary shapes,

$$s = \sqrt{\frac{2B\gamma V}{3(1-g)\Sigma}}, \tag{7}$$

where we accounted for the fact that $\omega_0 \to 1$ (a weakly absorbing scattering media limit). Equation (7) can be written as

$$s = \frac{1}{3}\sqrt{f\gamma d}, \tag{8}$$

where $f = B/(1-g)$ is the scaling constant. Therefore it follows that d can be determined from s if the scaling constant f is known:

$$d = \frac{9s^2}{\gamma f}, \tag{9}$$

where s is given by Eq. (5). Also we have $\sigma = 2\gamma f/3s^2\rho$. The complexity is due to the fact that the value of f has never been measured for snow, and the determination of d from s is heavily affected by the scaling constant f.

Theoretical considerations[9] point out that the scaling constant does not depend on the size but only on the shape and the real part of the refractive index of particles.[9] As a matter of fact, one can easily determine the product $d^* = fd$, but not d itself from Eq. (9). The theoretical estimations based on the ray-optics approach[9] show that $f = 11.5$ for spheres and $f = 7.4$ for fractal particles, which represent extremes never occurring in snow. One possibility to determine f for real-life situations is to measure R and d simultaneously. Then it follows that

$$f = \frac{9s^2}{\gamma d}, \tag{10}$$

or

$$f = \frac{9}{16\gamma d U^2(\vartheta_0,\vartheta,\varphi)} \ln^2\left[\frac{R_0(\vartheta_0,\vartheta,\varphi)}{R(\vartheta_0,\vartheta,\varphi)}\right], \tag{11}$$

where Eq. (5) was used. The measurement of R can be easily done. This is not the case for the effective grain size d because it involves measurements of both the surface area and the volume of grains. In particular, image analysis cannot be used to determine d because then only two-dimensional analysis is possible. This is the reason why the problem of the experimental determination of f was not formulated before.

We use the fact that SSA σ and, therefore, $d = 6/\rho\sigma$, can be measured by using CH_4 adsorption.[6,7,12] The principle of the method is to determine the number of methane molecules that can be adsorbed on the snow surface. In practice, the adsorption isotherm of methane on the snow must be recorded at 77 K. This is a lengthy operation requiring liquid nitrogen. Details of the techniques are given elsewhere.[12] Domine *et al.*[7] measured not only SSA but also the reflection function R at several wavelengths, the nadir observation, and incidence zenith angles in the range 64°–68°. This enables the determination of the scaling constant by using Eq. (11).

The dependence of the scaling constant f obtained from experimental data[7] using the technique outlined above is shown in Fig. 1. We assumed in theoretical calculations performed using Eq. (11) that $\lambda = 1.31\ \mu$m, $\vartheta_0 = 66°$, $\vartheta = 0°$ (as in the experiment[7]), and

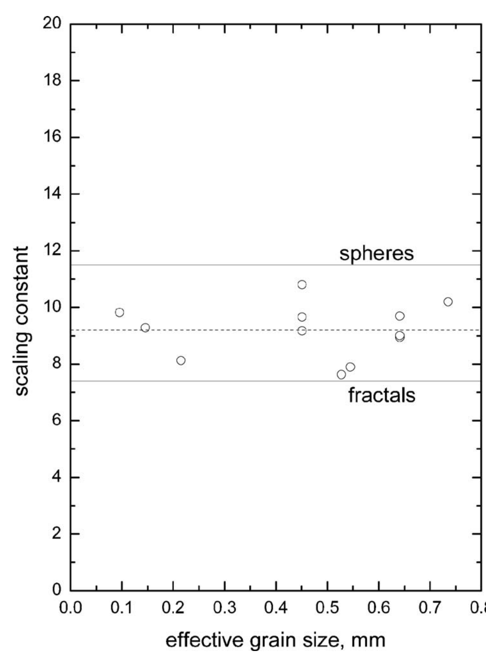

Fig. 1. Dependence of the scaling constant on the effective grain size. Solid lines give theoretical values of f for spheres and fractals. The dashed line corresponds to the value of the scaling constant equal to 9.2.

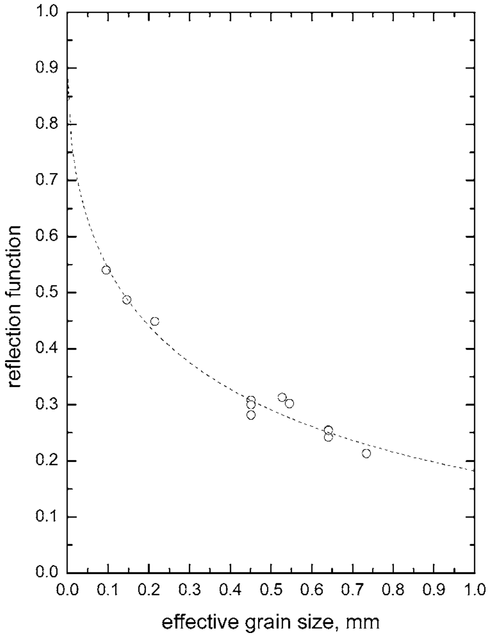

Fig. 2. Comparison of measurements (circles) and calculations (dashed curve). Further details are given in the text.

u_0 is given by Eq. (4). It follows that at this wavelength[13] $\chi = 0.0000132$. The value of $R_0 = 0.9$ for the geometry specified was found by using the radiative transfer code[8] for a semi-infinite turbid layer composed of fractal grains. It follows from Fig. 1 that the derived values of f are scattered between the theoretical values predicted for spherical and fractal ice grains. They also slightly depend on the grain size and the type of snow. Data shown in Fig. 1 suggest that the value of $f = 9.2$, which is close to the average of results for spheres and fractals, is representative for snow. It is interesting that although snow type is different for most of the measurements shown in Fig. 1, the value of f is not changed considerably. Therefore one derives the following simple approximate equation for the similarity parameter s [see Eq. (8)] in the case of natural snow as $s \to 0$:

$$s = \sqrt{\gamma d}. \tag{12}$$

This is a major result of this work.

The measured reflection function and the calculated one are presented in Fig. 2. Calculations have been performed by using Eqs. (1), (3), (4), and (12). It was assumed that $R_0 = 0.9$ as described above. This figure confirms that the derived scaling constant enables the interpretation of experimental results of Domine *et al.*[7]

The results obtained enable us to make the following conclusions. First, the value of the scaling constant determined in this work using experimental data[7] can be used to model the measured reflection function for various snow types applying Eqs. (1), (4), and (8). Second, the derived value of f in combination with Eqs. (9), (4), and (5) can be applied for the determination of the effective grain size[5] and the specific surface area,[7] avoiding time-demanding procedures such as optical microscopy and methane adsorption techniques. To our knowledge, the technique to determine SSA from measurements of the short-wave IR reflectance of snow was first introduced by Domine *et al.*[7]

All calculations require the value of R_0 for semi-infinite layers with irregularly shaped ice particles. R_0 can be found by using the code available on the Internet[8] or by using various approximations.[14] An attractive possibility is the measurement of the plane albedo[9,14] $r_p = \exp[-\sqrt{\gamma d} u_0(\vartheta_0)]$ or the spherical albedo[9,14] $r_s = \exp[-\sqrt{\gamma d}]$, where we used Eq. (12). Then calculations of R_0 are not needed. Also, measurements of R at two close wavelengths λ_1 and λ_2 in the near IR, where $\gamma(\lambda_1) \neq \gamma(\lambda_2)$, can be performed. Then the spectral dependence of R_0 can be neglected and bispectral measurements of the snow reflection function enable the simultaneous experimental determination of both d and R_0.

The approach presented above can also be applied to other weakly absorbing light scattering media with irregularly shaped particles.

The author (alexk@iup.physik.uni-bremen.de) is grateful to F. Domine for providing the results shown in Fig. 2 ahead of publication.[7] He thanks M. Mishchenko for the possibility of using the radiative transfer code based on the numerical solution of a nonlinear integral equation.[8]

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